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The twice renormalized rouse formalism of polymer dynamics. Segment diffusion, terminal relaxation, and nuclear spin-lattice relaxation

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Abstract

The twice renormalized Rouse formalism, a refined version of Schweizer's renormalized Rouse treatment of chain dynamics in entangled polymers, is presented. The time scale of validity is extended including terminal chain relaxation and center-of-mass diffusion. In clear contrast to the laws concluded from other polymer dynamics concepts such as the reptation (tube) model or the polymer mode-mode coupling formalism, the predictions perfectly compare with all results of recent spin-lattice relaxation dispersion and diffusion experiments as well as computer simulations. On the other hand, the twice renormalized Rouse formalism fails to explain the rubber-elastic plateau of stress relaxation. It is inferred that this is a consequence of the single-chain nature of the present approach not accounting for the fact that viscoelasticity largely is a manifestation of collective many-chain modes. In the rigorous sense, no such multi-chain treatment has been established so far to our knowledge. The necessity to consider inter-chain cooperativity in any really comprehensive polymer dynamics theory is concluded from low-frequency spin-lattice relaxation data, which are shown to reflect fluctuations of long-distance intermolecular dipole-dipole interactions.